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Historical Background of Ultrahigh Pressure Shock Compression Experiments at LLNL: 1973 to 2000

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I. Introduction

My purpose is to recount the historical development of ultrahigh pressure shock compression experiments at LLNL, which I experienced in the period 1973 to 2000. I used several experimental techniques: shock-impedance-match experiments using planar shock waves driven by nuclear explosives (NIMs), the Janus Laser, a railgun, and a two-stage light-gas gun.

Two things have motivated me: (i) the interaction between programmatic needs and scientific understanding (i.e., there are lots of scientifically interesting things to do here which are important programmatically) and (ii) accurate experimental data are required to develop accurate theoretical models (or as Feynman said, “If it (theory) disagrees with experiment, it is wrong”). The iteration between experiment and theory is commonly known as the scientific method.

I arrived at LLNL with a PhD in Condensed Matter Physics (thesis on measuring thermal and electrical conductivities of rare-earth single crystals), postdoctoral experience (measuring electrical and magnetic properties of Pu, Np, and U intermetallic compounds and alloys), three years experience teaching 7-8 different undergraduate physics courses while running the college’s computer terminal, a belief in the scientific method, and an intense interest in understanding materials physics. I managed to get one of the few jobs available in physics in 1973.

II. Shock Compression

My first assignment was in B Division where I learned about shock propagation in condensed matter. For example, I learned that thermodynamic states achieved depend on the structure of the shock front. In particular a series of reverberating shock waves produces substantially lower temperatures than produced by a single shock to the same final pressure. Years later I used a reverberating shock at the two-stage gun to achieve states well off the Hugoniot and to make the first observation of a metallic phase of hydrogen, something which had been searched for in the scientific community for ~100 years. In 1976 I moved to H Division to do experiments with Art Mitchell at the two-stage light-gas gun.

III. Janus Laser

When I moved to H Division I found that John Shaner, my new Group Leader, was building up a group to do EOS with laser-driven shock waves generated at the Janus Laser; a few experiments were also done at Argus. Jim Trainor was hired in 1977 to do this; Neil Holmes joined the laser effort in 1978. John Shaner moved to LANL in 1978 and I became Leader of the Shock Physics Group. These Janus laser experiments were extremely difficult. Shock transit times and distances were a few 100 ps and a few microns. The shock attenuated as much as a factor of ~2 in this time. Despite our best efforts, experiments could not be done with sufficient accuracy to affect theory, so I dropped our laser-driven EOS effort in 1981.

While I was unaware of the fact that the H Division laser-EOS program (5 people) was being used by LLNL as a selling point for the Laser Program, I quickly found out. I found myself several times in the office of Roy Woodruff, then AD for Defense Sciences (now called DNT). Hal Graboske was H Division Leader at that time and was instrumental in our making a smooth transition to two other ultrahigh pressure experimental methods: nuclear (shock) impedance match experiments (NIMs), which we felt we knew how to do, and the railgun, which needed development. Impact experiments, as with a railgun, are intrinsically more accurate than NIMs and railgun experiments in an LLNL laboratory could be done several times per week as opposed to one NIM per year at NTS.

IV. Nuclear Impedance Match Experiments (NIMs)

In 1974 I was put on a project to learn how to measure shock velocity in proximity to an underground nuclear explosion. Shock impedance match experiments involve measuring shock velocity in a pair of adjacent dissimilar materials. Thus, ultrahigh pressure EOS data could be measured if we could measure shock transit times across known thicknesses and if we could use one material whose EOS could be considered to be a standard. Measurement of shock transit times was initially difficult because a shock pin detector is voltage-charged and might experience ionizing gamma and neutron radiation prior to shock arrival. If this radiation causes the pin to discharge prior to shock arrival, the pin is useless. I designed radiation shielding for such pins. The shock pins and the detection system were designed and fielded by Art Mitchell. These pins and detection system were simply a more-robust version of the system Mitchell had implemented at the two-stage gun in B341. Eventually we learned how to measure shock velocities accurately downhole, which led to the NIM experiments.

Hal Graboske, then H Division Leader, and I went to talk to John Immele, then B Division Leader, and Seymour Sack about doing the first NIM. Immele said we could add our EOS package onto a certain device. I pointed out that this device had so much extraneous hardware that it might compromise generation of a planar shock front to impinge on our samples. Immele assured us that they knew what went on around a nuclear explosion and that we should accept their guidance, which I did. I designed the planar layers of the Al standard and samples. Mitchell and our technicians worked for nine months putting this package together in B341 and then in the field. Mitchell and I personally installed the package into the canister at NTS. When the shot was fired, no signals were measured on our fast-sweep oscilloscopes. The backup low-time-resolution recording system recorded signals much later. Thus, the shock generator did not work as promised. Mitchell anticipated a question that came up at the postmortem; namely, did the pin detectors have sufficient sensitivity to respond as expected. Mitchell did experiments at the two-stage gun that showed that the pins worked as expected at much lower shock pressures than at NTS, where they would be expected to work even better and did so on later experiments.

Being the tough and honest man that he was, Immele said that the next year the NIM would be the main experiment and the nuclear explosive would be the “add on”. Pat Crowley of A Division designed the planar shock-wave generator. Ray Heinle was in charge of interfacing our package with the L Division diagnostic system. I designed the EOS package. This NIM and the next two were successful. Once we learned how to do these experiments, Pu^{242} samples were put on the next two shots and the number of samples on both was doubled. Mitchell was assisted by Neil Holmes on the next two shots. Gordon Repp and Bob Tipton designed the shock generators on the next two shots. Pressures in Cu of 25, 15, and 7.8 Mbar were achieved in the three experiment. We qualified Al as the EOS standard by finding that John Moriarty, Marvin Ross, and Neil Ashcroft (Cornell U.) all calculated the same Al EOS. I analyzed the shock arrival times and determined the EOS data for Pu, U, Be, etc by shock impedance matching, along with calculating their error bars.[8] We published the unclassified Be NIM data only when it was in LLNL's interest to do so to show the good comparison with NOVA data.[11a]

After doing 4 of these shots Mitchell was tired and stressed; he also did experiments at the two-stage gun in B341. Since the technology had been thoroughly developed, I asked B Division to have someone from L Division do one NIM experiment per year. The decision was negative and the NIM series ended. Thus ended my twelve-year experience with classified experiments.

V. Railgun

The projectile in a railgun is accelerated by JxB forces in the plasma armature just behind the projectile, the so-called propulsive arc. The maximum velocity of the projectile is limited by the maximum velocity at which pressure can be transmitted in the driving medium to the projectile, which is the speed of sound in the driving medium. Since the speed of sound of an electromagnetic force is the speed of light, projectile velocity is limited, in principal, by the speed of light. This is substantially higher than the speed of sound in any light molecular gas, such as H_2. Thus, it was reasonable to try to accelerate projectiles in a railgun to obtain higher impact velocities, which in turn means higher shock pressures in the laboratory than with a gas gun.

We worked on a railgun project from 1981 to 1985.[12] Our goal was to obtain 12 km/s; we could get 8 km/s with our two-stage gun. A 50 % increase in impact velocity translates into a doubling of shock pressure. If we could get 12 km/s, then we would get the same pressures in the laboratory as in a NIM. Ron Hawke headed up our railgun project. We built two guns, 5 and 16 ft long, with Cu rails, a 1 cm diameter projectile weighing up to 4 g, and a He-gas injector which launched the projectile into the railgun at a velocity of 1 km/s. The projectile had a thin Al-foil armature at its rear which completed the electrical circuit across the rails when the projectile was injected into the railgun. The potential difference across the rails was established dynamically with a 625-kJ capacitor bank.

In this way, the Al arc did not dwell in one place very long to burn/damage the rails. All this was a substantial engineering accomplishment. Peak velocities of 3.0 of 6.6 km/s, respectively, were measured. Measured velocity agreed with our 1D hydro simulations up to a velocity of 4-5 km/s. At higher velocities the measured velocity was lower than predicted with a 1D code. Our diagnostics showed that additional arcs were struck near the breech which shunted current away from the propulsive arc. The source of the plasma which shunted the arc was Cu metal eroded from the rails at velocities above ~4 km/s. That is, a Cu layer 1 micron thick was eroded off the rails, as observed by measurements of inner diameter with a precision air gauge after a shot. John Nuckolls, Physics AD at the time, was a supporter of this project. However, Roy Woodruff, then AD for Defense Sciences, terminated the railgun project. The erosion problem is a materials problem which can probably be solved by plating a thin conducting layer of a refractory metal on the rails, such as Ta, W, or Mo.

VI. Two-Stage Light-Gas Gun

In 1976 I moved to H Division to do EOS experiments with Mitchell at the two-stage light-gas gun.[13] I had met Mitchell when I was working on experiments preliminary to the NIMs. I have never met another person who is so intensely driven to obtain voluminous amounts of highly accurate experimental data. He taught me a lot about how to do experimental physics. The two-stage gun achieves about twice the pressures achieved with plane-wave HE systems, which had been used extensively at LANL for EOS experiments.

A. Metals

Since in 1976 very few experiments had ever been done with a two-stage gun, we had to qualify metals with accurate Hugoniots to the highest available pressures. These metals are used as standards to shock compress other materials. For this reason Mitchell and I measured the Hugoniots of Al, Cu, and Ta and did the error analyses.[14] It was this experience which was so valuable in doing the NIMs. Having learned how to do this, in 1979-1980 we did U. Later, this method was applied to Pt.[17]

B. Molecular Fluids

From 1976 to 1992 we measured EOS states which were achieved primarily on the Hugoniot (single shock) and a few with double shocks. The quantities measured were Hugoniot EOS (P, V, E), temperature from emitted thermal radiation, electrical conductivities to get an idea of what charge carriers have a sufficiently large density to affect the EOS (and thus should be taken into account to develop a theoretical EOS). In one case, Raman spectra of water were measured to learn about the nature of inter-and intramolecular interactions which is necessary to know how to generate a theoretical EOS. Neil Holmes then did these nice experiments.

Ideas for experiments came from scientific discussions at conferences, other national laboratories, and universities. Going to conferences and other scientific exchanges is essential to drive scientific advancement. For example, the establishment of the Manhattan Project was driven by intense scientific interactions, most involving recent foreign-born immigrants to the US.[17a]

EOSs of dense fluids are important. The fact that a best theoretical estimate has been made does not make it correct. Knowledge comes from the iteration of experiment and theory. Any number of low-Z molecular materials have been, are and will be considered and/or used. This is especially true if there is an accurate EOS available when a design project starts. Useful EOSs of such materials cannot be measured during a project because, since EOSs of possible materials are not known, one is not sure which one would be best and by the time one is chosen by guess work, the measurements cannot be completed before the project is terminated. So I went to work on things that were and/or could be used. The flexibilty to do this has since been destroyed by the “Campaigns”.

Francis Ree et al of H Division develop EOSs of reacted explosives theoretically. To do so, interaction potentials between like molecules and algorithms to obtain interaction potentials between a mixture of dissimilar molecules are needed. Interaction potentials are derived from Hugoniot data. So I went to work measuring Hugoniots and double-shock points of several individual molecular products in reacted explosives, the most common being H_20[18-20], N_2[21-24], and CO_2[25]. CO[22,26], O_2[21,27], and air[25] were studied as well. Since hydrocarbons are commonly used as simulants of chemically reacting explosives, I collected data on many of these as well.[26,28,29] In order to understand relatively complex diatomic molecules, rare-gas liquids were studied to understand the basic interaction between simple monatomic fluids, such as Ar[21,30] and Xe[31] in which there are no vibrational excitations and electronic excitations are readily apparent in the data.

Since Hugoniot experiments have never been performed on binary mixtures of molecular liquids, the algorithms used to calculate effective pair potentials between the most abundant molecular species have never been tested experimentally. In 1989 I tried to measure Hugoniots of these binary mixtures. To do these experiments a new sample holder is necessary because the sample must inititially be at high static pressure and temperature to mix the two components on an atomic scale before shock compression. The Head of B Division's HE Program gave me funds to do this. However, to analyze results for a mixture, the EOS of each individual liquid must be known and that meant pure CO_2 needed to be done prior to doing the binary mixtures. I was told I could not do CO_2, only mixtures. I did CO_2 the first year because it was the scientifically correct thing to do; funding to do mixtures was then terminated. The CO_2 Hugoniot we measured shows chemical reactions at high pressures and temperatures. This work motivated the recent LLNL discovery of new phases of CO_2 in a laser-heated diamond cell at high pressures. Similarly, molecular dissociation I observed in fluid N_2 in the mid 1980s led to the prediction by H Division theorists that diatomic N_2 would transform to polymeric N at room temperature and Mbar pressures. A recent report in 2000 claims to have observed this in a DAC. We did measure a mixture of water, ammonia, and isopropanol because these molecular liquids mix at ambient pressure and temperature.[32-34]

Hydrogen in the form of D/T is the fuel in ICF. For this reason I began designing liquid-H_2 cryogenic (T_0=20 K) sample holders in 1976. Initially we did less cryogenically challenging liquids (T_0=80 K)[21] to get an idea of the difficulties both in theoretically analyzing the data and with the experiments. We then measured single and double-shock Hugoniot and temperature points and electrical conductivities under single-shock compression of liquid D_2/H_2.[35-37] Corresponding Hugoniot data of liquid He (T_0=4 K)[38] was then measured.

Marvin Ross' theoretical analyses of our shock data was crucial in keeping our shock program going for decades.

In 1992 we used H_2 and D_2 to achieve states well off the Hugoniot along a quasi-isentrope up to a pressure and compression 10 and 3 times greater than achieved on the Hugoniot (single shock).[39-41] These states are significantly off the principal Hugoniot and were achieved by using a reverberating shock wave. Since LLNL ICF targets go through this regime and previous shock experiments were done primarily on the Hugoniot, this is a major technological advance. Since we measured electrical conductivities and we found metallic fluid hydrogen, this generated intense scientific interest.[42] The quest for a metallic phase of hydrogen had been going on for ~100 years. These experiments grew out of the fact that we had been doing measurements of D_2 electrical conducucuctivities on the Hugoniot (single shock) and we also had been using a reverberating shock both to compact high-T_c oxide powders and to shock-synthesize novel phases of C. It took from 1992 until 2000 to show that the electrical conductivities of shock-reverberated H_2, O_2, and N_2 are systemically similar to each other with electronic charge carriers and water is systematically dissimilar with proton charge carriers.

C. Planetary Fluids

The giant planets Jupiter, Saturn, Uranus, and Neptune consist of molecular fluids at high P and T. They are effectively big balls of reacted explosives. Thus, data we took on molecular fluids were also applied to developing pictures of the interiors of giant planets.[32-34,43-46] FeO was studied because of its likely presence at the core-mantle boundary in the Earth.[47]

D. Shock-Wave Profiles

Measuring shock-wave profiles in solids is important for understanding dynamic strength and phase transitions. Because Sandia was doing excellent work on dynamic strength and had the expertise and diagnostics in place to do it, in 1982 I initiated the formal request from LLNL to Sandia to perform such experiments. Sandia did an excellent job on the Be strength experiments. Our group had just spent 6 years and several $M to develop the techniques to be able to do cryogenic fluids. It didn't make sense for us to throw all that time and money away to do something that Sandia could do in a few months.

Eventually we did develop a VISAR to do wave-profile measurements. Dave Erskine and I used a VISAR to observe the graphite-diamond transition[48] and applied it to tuff from NTS.[49]

E. Materials Recovered from High Shock Pressures

While running hydrocodes in the mid 70s, I was struck by the ultrahigh pressures (~Mbar), temperatures (few 1000 K), and quench rates (10^{12} bar/s and 10^{9} K/s) which could be achieved in the laboratory with gas guns and explosives. If materials could be recovered intact from these extreme conditions, they might have interesting structures and properties. When Claire Max asked me in 1984 to headup the Center for High Pressure Sciences in IGPP, I realized that it was an opportunity to try to learn how to recover materials from extreme dynamic conditions. On suggesting collaborations in this area to university professors, I found only enthusiasm. Physicists like to try new things. So I worked on shock recovery experiments for ~10 years with the goal of doing things which had not been done previously.[50,51] Claire helped me get a small two-stage gun built in an unclassified area and supported it for IGPP collaborations with UC researchers. Grad students were supported with IGPP grants.

These recovery experiments showed that many things are possible with shock waves which were not thought to be so previously. These results might enable new technologies; e.g., issues connected with Stockpile Stewardship.

My areas of interest were: (i) investigating shock-induced melting and rapid resolidification,[52] (ii) recovering samples from the highest possible pressures because this would cause the widest range of possibilties (1.2 Mbar is our max[53,54]), (iii) using the thinnest samples because this would cause the highest quench rates and likelihood of retaining high-pressure effects (1 micron from 1.0 Mbar is our thinnest[55,56]), (iv) synthesizing nanocrystalline materials,[57] (v) synthesizing metastable materials,[58-60] (vi) synthesizing carbon phases from C-60,[61-63] (vii) inducing enormous densities of shock-induced defects which alter physical properties (superconductors have higher critical current densities[64-68] and permanent magnets are more permanent[69]), (viii) crystallographically oriented shock compacts,[70] (ix) shocking oriented crystals at an angle to the impactor to maximize defect generation without fracturing the sample,[71] (x) shock compacting ceramic powders to get high densities with microstructural features which would enhance performance,[72-75] (xi) shock compaction of metastable materials to make consolidated powder compacts by working completely out of the equilibrium phase diagram,[76] (xii) comparing experimental data with a computer code which simulates shock compaction of powder particles,[77,78]

(xiii) shock-induced amorphization,[79,80] and (xiv) shock-induced geophysical phenomena.[81,82]

VII. Postdocs

Over the past 10 years I've had the pleasure of working with several postdocs on this work: Andy Gratz, Peter Fiske, Sam Weir, Ricky Chau, and Marina Bastea.

VIII. Teller Fellowship

In 2000 I was awarded one of the first Edward Teller Fellowships. I'm proud to have received this because Edward Teller epitomizes the combination of strong science in the cause of national defense.[17a]

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